Single-molecule conductivity of non-redox and redox molecules at pure and gold-mined Au(111)-electrode surfaces

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The structure, two-dimensional organization, and function of molecules immobilized on solid surfaces can be addressed in a degree of detail that has reached the level of the single-molecule. In this context redox molecules are "smart" molecules adding sophisticated electronic function. Redox metalloproteins such as blue copper, heme, and iron-sulfur proteins as well as redox metalloenzymes are other new targets for single-molecule electrochemical and bioelectrochemical surface science. Electrochemistry combined with scanning tunnelling and atomic force microscopy in aqueous chemical or biological media supported by comprehensive theoretical frames, have emerged as core approaches in these exciting areas.

Single-molecule redox electrochemistry is rooted in two major areas. One is the preparation of well-defined (atomically planar) electrode surfaces modified by molecular monolayers (SAMs). High-resolution in situ STM combined with large-scale theoretical computations have offered detailed insight in the surface binding modes of functionalized alkanethiols on Au-surfaces in particular. Such surfaces have disclosed a variety of surface structures that involve both direct binding to a planar Au and binding to surface-mined Au-atoms. In addition the SAMs ensure protein/enzyme immobilization gentle enough that the proteins retain electron transfer or enzyme activity in a variety of local environments. The second area is the mapping and control of the immobilized redox molecules and metalloproteins themselves. Single-molecule resolution has also here been achieved, both in structural mapping and in controlled single-molecule electron transport and enzyme function on SAM surfaces such as those now characterized to (sub)-molecular resolution.

Three recent references:

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